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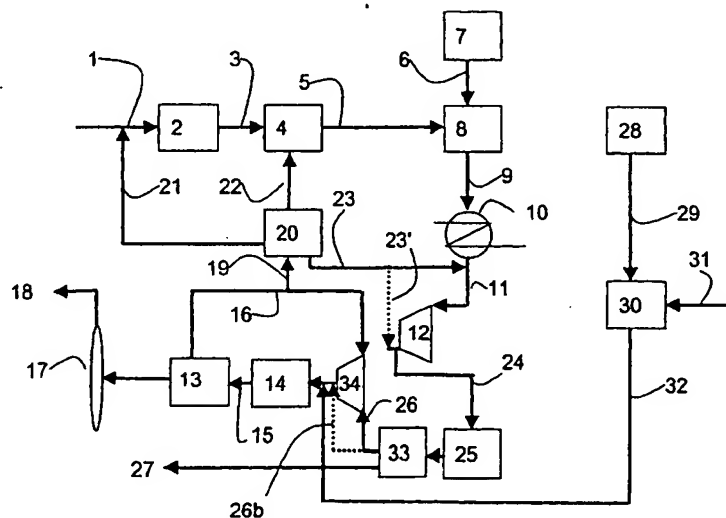
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[Continued on next page]

(54) Title: METHOD AND PLANT FOR PRODUCTION OF OXYGENATED HYDROCARBONS



(57) Abstract: A method for increasing production in an existing processing plant for converting natural gas into a product, wherein the natural gas is first converted into a synthesis gas in a synthesis gas section, the synthesis gas is reacted in a reactor for synthesis of the product, where non-converted synthesis gas and product are separated into two streams, where a product-rich stream is taken out of the process, whilst a product-poor stream is recycled as feed to the reactor together with make-up synthesis gas, and where a portion of the recycle stream is taken out of the recycle loop as a purge gas, where the purge gas is separated into hydrogen-rich and hydrogen-poor streams, where hydrogen-rich streams are introduced into steps in the process where it is desirable to have a supplement of hydrogen, and where the residual thermal value of the hydrogen-poor stream is optionally used for heating before it is discharged. A modified processing plant for carrying out the method is also described.

WO 01/47846 A1



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METHOD AND PLANT FOR PRODUCTION OF OXYGENATED HYDROCARBONS

The present invention relates to a method for increasing production in an existing
5 processing plant and to a processing plant that has been modified in order to carry out the method.

Today, in the building of new processing plants, such as, e.g., plants for the production of methanol from natural gas or other suitable carbon sources, there is a clear trend
10 towards building plants with an ever-increasing production capacity, such as more than 5000 tonnes of methanol each day. Thus, production costs are reduced due to economies of scale.

In areas where the price of natural gas is low, it is possible to produce methanol at a cost
15 of as little as about USD 80 per tonne, and this is allowing methanol to obtain a foothold in a fuel market, i.e., for fuel cell cars and electricity production.

For existing processing plants that are too small in relation to current demands, there is a need to find solutions which allow an increase of total production and thus a reduction
20 in production costs per unit without having to make major costly conversions and refits of the existing processing plant.

It is thus an object of the present invention to provide a method of increasing production in an existing processing plant without necessitating major, costly conversions of the
25 existing plant. and to provide a processing plant that has been modified in order to carry out the present method.

According to the present invention, this is achieved by means of a method for increasing production in an existing processing plant for the conversion of natural gas to a product,
30 wherein the natural gas is first converted to a synthesis gas in a synthesis gas section, the synthesis gas is reacted in a reactor for synthesis of the product, where non-converted synthesis gas and product are separated into two streams, where a product-rich stream is taken out of the process. whilst a product-poor stream is recycled as feed

to the reactor together with make-up synthesis gas, and where a portion of the recycle stream is taken out of the recycle loop as a purge gas, where the purge gas is separated into hydrogen-rich and hydrogen-poor streams, where hydrogen-rich streams are introduced into steps in the process in which it is desirable to have a supplement of hydrogen, and where the residual thermal value of the hydrogen-poor stream may, if
5 desired, be used for heating before it is discharged.

It is preferable that the synthesis gas from the synthesis gas section should be fed with a hydrogen-rich stream from the separated purge gas, and that this hydrogen-enriched
10 synthesis gas should be passed through a new once-through reactor for production of the product and through a unit in order to separate a product-rich stream that is taken out, and a product-poor stream that is used as feed in the original reactor.

It is also preferable that the product-poor stream which is used as feed for the original
15 reactor should be fed with additional synthesis gas that is produced in a separate secondary synthesis gas line.

The existing reactor will preferably be operated virtually unchanged.

20 It is preferable that the secondary synthesis gas line should be based on ATR or POX.

It is also preferable that the product should be methanol or dimethyl ether.

Also provided is a processing plant for the production of a product based on natural gas,
25 where the processing plant comprises a synthesis gas section for producing synthesis gas that mainly consists of CO, CO₂, H₂ and water, a synthesis section where the product is formed, and a purification section where the product is separated from unconverted reactants and other substances and purified, where unconverted reactants that are separated from the product are recycled to the synthesis section, and where a
30 portion of the gas that is recycled is drawn off in a line in order to prevent build-up of inert gases, where the plant also comprises a separation unit for separating the gas that is drawn off in a line into hydrogen-rich and hydrogen-poor fractions, a line for conveying hydrogen-poor fractions to optional combustion and lines for leading

hydrogen-rich fractions into the natural gas feed and for recycling to the synthesis section respectively.

It is preferable that between the synthesis gas section and the synthesis section there
5 should be provided a once-through reactor for synthesis of the product, and a separation unit for the separation of a product-rich fraction to a line and a product-poor fraction to a line leading to the synthesis section.

Furthermore, it is preferable that the processing plant should also comprise a separate
10 secondary synthesis gas line for producing a secondary synthesis gas, and a line for conveying the secondary synthesis gas as feed for the synthesis section.

The invention will be described below with the aid an example and the attached figures, wherein:

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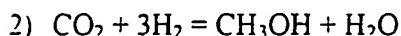
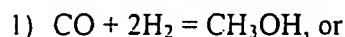
Figure 1 is a schematic illustration of the structure of a traditional plant for the production of methanol from natural gas; and

Figure 2 is a schematic illustration of a plant according to the present invention.

20 Today, the production of methanol is carried out essentially as shown in the schematic diagram in Figure 1. The processing plant consists basically of three sections, a synthesis gas section 2, 4, 8) where production is normally based on natural gas (NG), a synthesis section 14 where the actual methanol synthesis takes place, and a distillation section 17 where the methanol produced is purified.

25

The methanol synthesis is takes place by means of the two following reactions:



30 The synthesis gases, which essentially comprise CO, CO₂ and H₂, in addition to water and non-reacted hydrocarbons. are prepared following one of three different concepts. namely:

a) conventional steam reforming,

- b) conventional autothermal reforming with a catalyst (ATR) or without a catalyst (POX), or
- c) a combination of a) and b).

5 Before the natural gas is sent in line 1 to the reformer for synthesis gas production, sulphur compounds are removed in a conventional manner, and steam is then saturated in and/or added directly to the gas. Saturation can also take place by using a so-called "saturator". Normally, the gas is also treated in a so-called pre-reformer 2 before it is sent into the reformer 4, 5, in order to convert all higher hydrocarbons.

10

The following chemical reactions take place during the production of synthesis gas:

- 3. $\text{CH}_3 + \text{H}_2\text{O} = \text{CO} + 3\text{H}_2$, steam reforming
- 4. $\text{CH}_4 + 1.5 \text{O}_2 = \text{CO} + 2 \text{H}_2\text{O}$, partial oxidation
- 5. $\text{CO} + \text{H}_2\text{O} = \text{CO}_2 + \text{H}_2$, shift reaction

15

Reactions 3 and 5 in the reforming reactor are highly endothermic and the heat that is necessary for the reaction can either be supplied by external firing, as in a steam reformer, or by means of a combination with partial oxidation, according to reaction 4, as in an autothermal reformer.

20

In a steam reformer (SR), natural gas (NG) (methane) is converted in a tubular reactor at a high temperature and a relatively low pressure. A traditional steam reformer consists of a large number of reactor tubes, usually 100 to 1000, having a tube length of 10 – 16 metres, where each tube has an internal diameter of about 10 cm and an external
25 diameter of about 12 cm. This unit may have a length of as much as 50 metres, a width of more than 10 metres and a height of more than 20 metres, with the result that such a reactor will require a relatively large space.

Conventional steam reformers are operated in a pressure range of from about 15 to 40
30 bar. The outlet temperature from such a reformer may be as much as 950 °C. The heat required to operate the reaction is supplied by means of external firing or heating and the reformer can be top, bottom or terrace-fired. The heat can also be transferred to the reaction by means of convective heat as in a heat exchange reactor. The ratio of steam

to carbon is from 1.6 to 4 and the ratio of H₂ to CO in the product stream from the reformer is about 3. A typical synthesis gas from a conventional steam reformer contains about 3 volume % methane.

5 In an autothermal reformer (ATR), synthesis gas production is carried out mainly by means of reactions 3 and 4, so that the heat necessary for reaction 3 is generated internally by reaction 4. In an ATR, natural gas (methane) is brought together with oxygen-containing gas such as, for example, air, inside a combustion chamber. The temperature in the combustion chamber may rise to more than 2000 °C. After the
10 combustion, the reactions are brought into equilibrium by means of a catalyst before the gases exit the reformer at a temperature of about 1000 °C. The size of an ATR may be a height of 10 – 20 metres and a diameter of about 4 – 7 metres.

An alternative autothermal reformer uses a concept called partial oxidation (POX). A
15 reformer of this kind does not contain a catalyst to expedite the reactions and therefore as a rule will have larger dimensions than an ATR.

The reforming of natural gas can also be effected by combined reforming (CR), where the reformer section consists of an SR and an ATR. A combination of SR and ATR
20 makes it possible to adjust the composition exiting the reformer section by controlling the admissions of the two reformers. In CR, SR is operated under milder conditions than in normal SR, i.e., at a slightly lower temperature. This results in a slightly higher methane content in the gas released from the reformer. This methane content is converted in the subsequent ATR. The ratio of carbon to steam in a reformer of this
25 kind is in the range of 1.2 to 2.4, with a ratio of hydrogen to CO in the product gas of well over 2. The optimal stoichiometric number
($SN = (H_2 - CO_2) / (CO_2 + CO)$) for methanol synthesis is about 2.05.

Figure 1 shows a synthesis gas section of the CR type. However, it is not critical what
30 type of synthesis gas section is included in the plant. A plant equipped with a synthesis gas section of the ATR type will not have SR 4, whereas a plant of the SR type will not have an ATR 8 and air separation unit 7 with accompanying line 6.

After the synthesis gas section 2, 4, 9, the synthesis gas is conveyed in line 9 to a heat exchanger 10 where it is cooled. After the heat exchanger 10, the synthesis gas is conveyed in line 11 to a compressor 12 where it is compressed to the desired pressure in the methanol synthesis section, which is typically about 80 bar.

The methanol synthesis in the synthesis section takes place according to reaction equations 1 and 2 above, and is an exothermic process where conventionally several different types of reactors 14 are used, such as:

- An isothermal tubular reactor with catalyst on the inside of vertical tubes and boiling water on the outside. The reaction heat will be removed by partial evaporation of the water.
- Adiabatic fixed bed reactors with cooling between each step
- A fluidised bed reactor
- Adiabatic reactors with cooling by means of a supply of new feed at several levels downwards in the reactor (quench converter system).

After the reactor 14, the product is fed via a line 15 to a crude methanol separator 13 that separates the product stream into a methanol-rich stream 34 and a methanol-poor stream 16. The methanol-rich stream in line 34 is fed to a conventional methanol purification unit 17 that sends methanol out in line 18.

The methanol-poor product is usually led via a recycle line 16 back to the reactor 14. Alternatively, the reactor is a once-through reactor without recirculation that can be followed by one or more similar reactors placed in series.

A synthesis loop 13, 14, 15 and 16 is shown in Figure 1. The actual recycle loop consists of a heat exchanger ("inter exchanger") (not shown) that preheats the feed to the synthesis reactor and cools the production gas, the synthesis reactor(s) 14, a crude methanol separator 13 and a system for recovering energy from the exothermic methanol synthesis reactions (not shown).

A so-called purge stream is taken out of this recycle loop via line 19 to prevent the accumulation of inert (non-reacting) gases in the recycle loop. The purge gas in line 19

is often split into a first stream 19a that is conveyed together with the feed gas in the gas supply 1 and a second stream 19b that is used as combustion gas for heat-consuming processes in the methanol synthesis, such as in the steam reformer 4, or in another process at the same plant, or is discharged.

5

One of the problems of the prior art is that inert gases, primarily nitrogen, in the recycled purge gas will react with other constituents of the synthesis gas and be converted to NO_x and NH_3 at the high temperatures that prevail in the reforming plant. According to the present invention, this is avoided in that the portion of the purge gas
10 that is rich in inert gas is used only as fuel for energy-consuming processes, such as the steam reformer 4.

Figure 2 illustrates a preferred embodiment of the present invention which has been based upon a traditional existing methanol synthesis plant as described above. A plant
15 having an existing reformer section based on CR has been taken as the basis.

In this embodiment, purge gas that is taken out of the recycle loop in line 19 is separated in a separation unit 20 into three streams, two hydrogen-rich gas-streams 21, 23 at different pressures, and a hydrogen-poor stream 22. The separation unit 20 is a
20 conventional hydrogen recovery unit which either works according to the Pressure Swing Adsorption (PSA) principle, the membrane principle or is of a cryogenic type. The hydrogen-rich fractions preferably have a hydrogen content of 70 to 100 %.

One of the hydrogen-rich streams 21 is fed into the gas supply 1 and is mixed with
25 incoming natural gas. The other hydrogen-rich stream 23 is led into the synthesis gas stream in line 11. Alternatively, depending upon the relative pressures in the different parts, this hydrogen-rich stream 23 is led via line 23' into the synthesis gas stream after the compressor 12 inside a line 24. The hydrogen-poor stream 22 is sent to the burners in the steam reformer 4 as fuel.

30

Line 24 conveys the synthesis gas stream from the compressor 12 to an extra once-through reactor 25 for the production of methanol. This synthesis gas stream is enriched with hydrogen from line 23 or 23'. The reactor 25 will normally give a yield of about

30% methanol, that is to say that about 30 – 35 % of the carbon entering the reactor is converted to methanol. The product stream from the reactor 25 is separated in a crude methanol separator 33 into a methanol-rich stream 27 and a stream 26 that consists primarily of non-reacted synthesis gas and inert gases from the methanol synthesis.

Stream 26 is led into a recycle compressor 34 in the recycle loop 16 and is introduced into the existing reactor 14. Alternatively, if the pressure in line 26 is or is set sufficiently high, stream 26 can be fed as indicated at 26b past compressor 34 and directly to the recycle loop and reactor 14.

The stoichiometric number, SN, for the synthesis gas in line 9 will normally be about 2.06, whilst the SN in line 24 will normally be greater than 2.06 because of the supply of hydrogen-rich gas from line 23 or 23'. The methanol-poor stream 26 has a high hydrogen content in relation to other reactive gases, i.e... a high SN that normally will be greater than 2.10.

In order to reduce the SN of the gas from line 26, the recycle line 16 is also fed with a synthesis gas having a lower SN produced in a separate secondary synthesis gas line 28, 29, 30, 31, 32. This secondary synthesis gas line comprises an ATR or POX reactor 30 which receives oxygen as almost pure O₂, oxygen- enriched air or air, via line 29 from the oxygen unit 28 and natural gas from line 31. This new synthesis gas is fed to the recycle line 16 through line 32. This secondary synthesis gas line could also include non-illustrated units such as a compressor and heat exchanger etc. The gas that is introduced into the reactor 30 is pre-treated in the same way as the original synthesis gas line, 2, 4, 8. Surplus heat in the process can be used in the different separation steps: if desired, the natural gas feed to ATR or POX 30 can be heated by using an extra fire heater or heat exchanger using hot synthesis gas from ATR or POX 30.

A combination of preheating and steam reforming in a convective reformer before ATR or POX 30 is also conceivable.

Here, it is important that the new separate ATR or POX reactor 30, which produces a synthesis gas having an SN of less than 2, has a capacity sufficient to allow the SN of

the feed to the synthesis reactor 14 to be reduced to about 2.06 from an SN of more than 2.10 in the gas in line 26.

The once-through reactor 25 is usually a once-through reactor of the simplest possible type, i.e., of the isothermal tubular reactor type as mentioned above with reference to reactor 14, and will on the outside of the tubes of the reactor produce more than enough steam at the right pressure to obtain a favourable steam to carbon ratio inside the reformer. Normally, 30 to 40% of the steam generated on the outside of the tubes in the reactor 25 for the cooling thereof will be used as feed for the new reformer 30 in order to obtain the desired steam/carbon ratio therein. The rest of the steam can be used in other steam and/or heat consuming processes, such as, for example, for purifying / further processing of the methanol stream. Typically, the methanol-rich product stream contains about 15% water and some ethanol, and distillation is usually carried out in order to obtain pure methanol.

The conversion in the new reactor 25 will affect the conversion in the existing synthesis reactor 14 so that a reduced conversion in reactor 25 will lead to more unconverted synthesis gas being fed from reactor 25 to the synthesis reactor 14, with the result that there will be an increase in the methanol production in 14.

The amount of the gas in the recycle line 16 that is taken out as purge gas through line 19 and the amount that is recycled in line 16 directly to the synthesis reactor 14 can be varied in order to optimise the system.

Examples

The table below shows simulation results for two examples, one for an existing methanol process according to the outline in Figure 1, and one for a plant according to the present invention.

The basis for the simulation is natural gas having a methane content of about 82% methane. The oxygen feed is varied so that the CH_4 slip is about 1.36%. The admissions in the existing reformer section are the same in both examples.

The new ATR operates at 35 bar and the steam to carbon ratio (S/C) to the pre-reformer in the new line II is 1.0. The once-through reactor 25 is positioned after the compressor 12 and has an outlet pressure of 80 bar.

		Example 1	Example 2
		Existing methanol process	New concept for increased methanol production
Line I			
NG-rate to existing line	Normalised (%)	100	100
NG-fuel	%	100	100
Oxygen to secondary reformer 8	%	100	110
S/C in feed to reformers 4, 8	%	1.8	1.8
Temperature in primary reformer 4			
Duty	%	100	100
Temperature in secondary reformer 8	°C	1000	1000
Line II			
NG-rate to ATR line 31	% in relation to existing plant		26
S/C feed to new pre-reformer			1
Oxygen to new ATR 30	% in relation to existing plant		40
Inlet temperature in new ATR 30	°C		600
Outlet temperature in new ATR 30	°C		1000
Outlet pressure from new ATR 30	bar		35
CH ₄ slip from new ATR 30	mol%		1.4
Stoichiometric number			
SN 1. line I from pre-reactor			2.13
SN 2. line I, feed pre-reactor			2.08
SN 4. line II from new ATR 30			1.68
SN from CR, line 9			2.02
SN 3. line I+II, MUG to			2.06

synthesis loop			
Synthesis loop			
Recycle ratio in synthesis loop		4.2	4.2
Total consumption and production			
Total NG to main process	%	100	
Total O ₂ -consumption	%	100	
Total production of crude methanol, I+II (methanol content in the crude methanol)	%	100	126

As can be seen from the table above, it is possible in the illustrated example to substantially increase the production of crude methanol in a process plant for methanol production, without subjecting the original plant to more stresses than during traditional operations. This is primarily of importance for extensions of existing plants where it is desirable to use the existing plant to the greatest extent possible, without having to redimension and rebuild large parts of the existing plant.

For the modified plant, it is possible to use surplus materials or unconverted amounts of a type of reactants in the process by supplying new amounts of other reactants and thus increase production, thereby rendering it less expensive.

It is important to note that the layout of the individual modules and constituents in the exemplified plants may differ from that which initially can be understood from the figures. Elements that the skilled person knows are included or can be included in such plants, such as heat exchangers, compressors, pressure-relief tanks etc., have to some extent been omitted as they are of no significance to the invention. Similarly, the assembly of some of the elements can differ. Thus, some of the elements that are drawn as one unit may consist of several similar or dissimilar elements connected in series and/or in parallel relation. For instance, the reactor 14 may comprise a plurality of parallel-connected and/or series-connected reactors.

The present method and process plant are also useful in connection with the extension of plants for the production of oxygenated hydrocarbons other than methanol, such as,

for example, dimethyl ether. The construction and mode of operation of a plant for the production of dimethyl ether are quite similar to those of a plant for methanol production, and thus the problems are completely parallel. Although the invention has been described with reference to a plant for the production of methanol, it also comprises other plants as mentioned above.

P a t e n t c l a i m s

1.

A method for increasing production in an existing process plant for converting natural
gas to a product, wherein the natural gas is first converted into a synthesis gas in a
synthesis gas section, the synthesis gas is reacted in a reactor for synthesis of the
product. where non-converted synthesis gas and product are separated into two streams,
where a product-rich stream is taken out of the process, whilst a product-poor stream is
recycled as feed to the reactor together with make-up synthesis gas, and where a portion
of the recycle stream is taken out of the recycle loop as a purge gas,
c h a r a c t e r i s e d i n that the purge gas is separated into
hydrogen-rich and hydrogen-poor streams, where the hydrogen-rich stream is
introduced into steps in the process where it is desirable to have a supplement of
hydrogen. and where the residual thermal value in the hydrogen-poor stream optionally
is used for heating before it is discharged.

2.

A method according to claim 1, c h a r a c t e r i s e d i n that
the synthesis gas from the synthesis gas section is fed with a hydrogen-rich stream from
the separated purge gas and that this hydrogen-enriched synthesis gas is passed through
a new once-through reactor for production of the product and through a unit in order to
separate a product-rich stream that is taken out, and a product-poor stream that is used
as feed in the original reactor.

3.

A method according to claim 2. c h a r a c t e r i s e d i n that
the product-poor stream that is used as feed for the original reactor is fed with additional
synthesis gas which is produced in a separate secondary synthesis gas line.

4.

A method according to one or more of the preceding claims,
c h a r a c t e r i s e d i n that the existing reactor is operated
virtually unchanged.

5.

A method according to one or more of the preceding claims,

c h a r a c t e r i s e d i n - that the new synthesis gas line is based
5 on ATR or POX.

6.

A method according to one or more of the preceding claims,

10 c h a r a c t e r i s e d i n that the product is methanol or dimethyl
ether.

7.

A processing plant for producing a product on the basis of natural gas, where the
15 processing plant comprises a synthesis gas section (2, 3, 4, 5, 6, 7, 8) for producing
synthesis gas that essentially consists of CO, CO₂, H₂ and water, a synthesis section (14)
where the product is formed, and a purification section (13, 17) where the product is
separated from unconverted reactants and other substances and is purified, where
unconverted reactants that are separated from the product are recycled to the synthesis
20 section (14), and where a portion of the gas that is recycled is drawn off in a line (19) in
order to prevent a build-up of inert gases, c h a r a c t e r i s e d
i n that the plant also comprises a separation unit (20) for separating the gas that
is drawn off in line (19) into hydrogen-rich and hydrogen-poor fractions, a line (22) for
conveying hydrogen-poor fractions to optional combustion, and lines (21, 23) for
25 leading hydrogen-rich fractions into the natural gas feed and for recycling to the
synthesis section (14) respectively.

8.

A processing plant according to claim 7, c h a r a c t e r i s e d i n
30 that between the synthesis gas section (2, 4, 8) and the synthesis section (14) there is
provided a once-through reactor (25) for synthesis of the product, and a separation unit
(33) for separation of a product-rich fraction to a line (27) and a product-poor fraction to
a line (26, 26b) that leads to the synthesis section (14).

9.

A processing plant according to claim 8. c h a r a c t e r i s e d i n
that it also comprises a separate secondary synthesis gas line (28, 29, 31, 30) for
s producing a secondary synthesis gas, and a line (32) to lead the secondary synthesis gas
as feed for the synthesis section (14).

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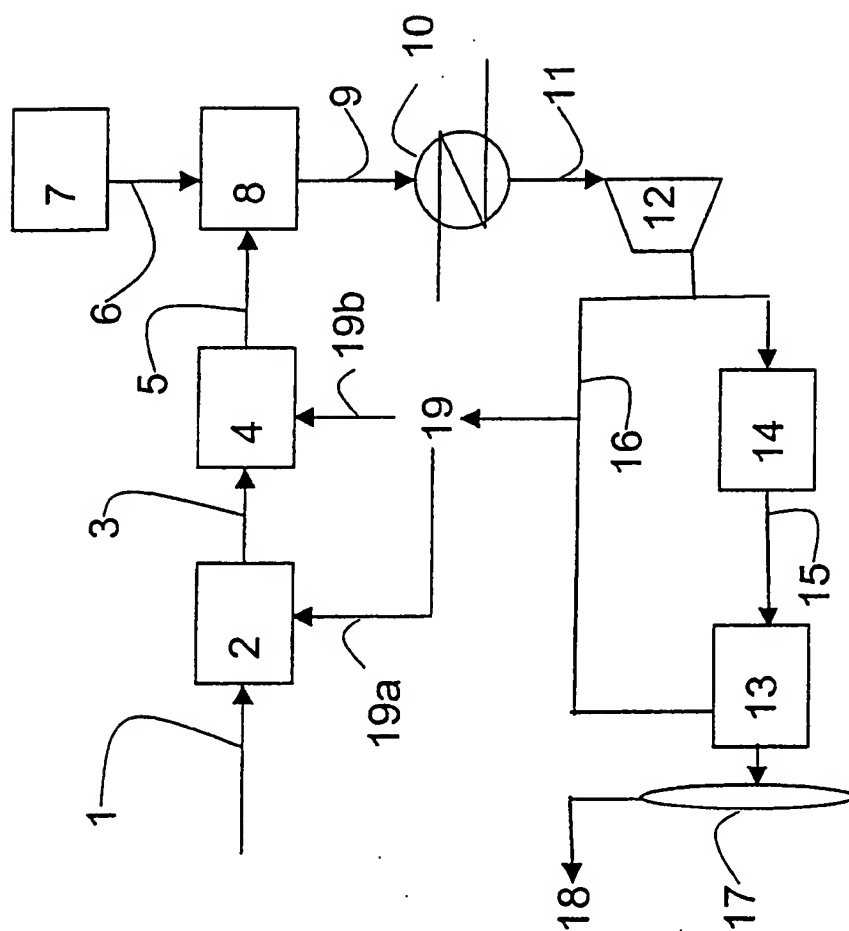


Fig. 1

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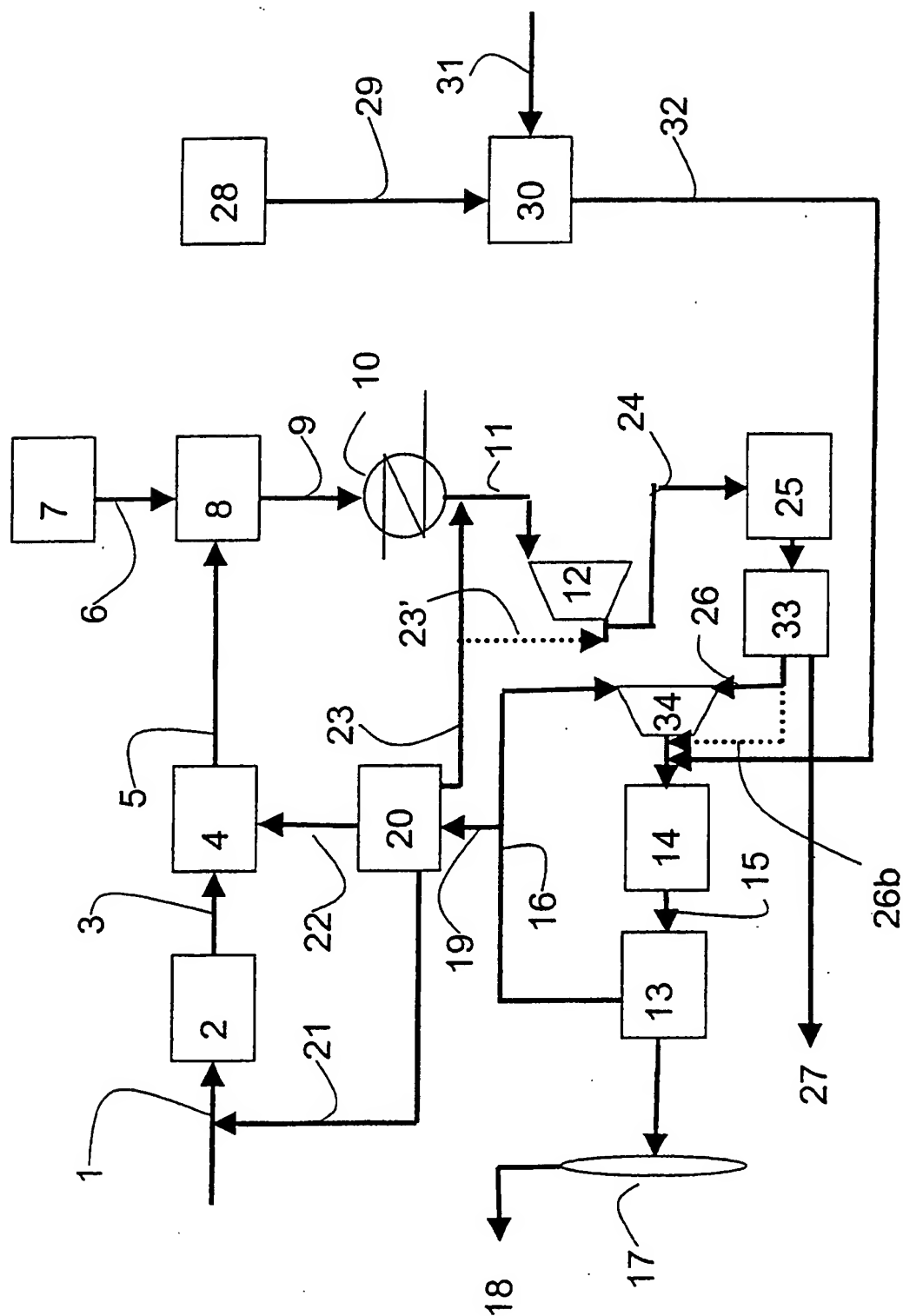


Fig. 2

INTERNATIONAL SEARCH REPORT

International Application No

PCT/NO 00/00450

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C07C29/151 C07C41/01

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C07C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EP0-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 233 076 A (FOSTER WHEELER ENERGY LTD) 19 August 1987 (1987-08-19) page 11, line 6 - line 14 abstract; claims 1-3 ---	1-9
A	EP 0 195 200 A (UHDE GMBH) 24 September 1986 (1986-09-24) the whole document ---	1-9
A	EP 0 123 534 A (FOSTER WHEELER ENERGY LTD) 31 October 1984 (1984-10-31) the whole document ---	1-9
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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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